Non-classical rotational inertia in the supersolid state

Shun-ichiro Koh
Physics Division, Faculty of Education, Kochi University
Akebono-cho, 2-5-1, Kochi, 780, Japan *

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An abrupt drop in the moment of inertia recently found in solid helium 4 is explained in terms of dynamics of zero-point vacancies (ZPV). Mechanical decoupling of ZPV from the motion of the container due to Bose statistics is developed to a macroscopic phenomenon by repulsive interaction. It gives a negative answer to the question whether BEC is a necessary condition for non-classical rotational inertia in a bulk three-dimensional system.

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Solid helium 4 has been termed a quantum crystal. On introduction of defects, it displays several anomalies not observed in classical crystals. Vacancies can move about unusually rapidly. Hence, it is natural to expect that the quantum-mechanical fluctuation delocalizes vacancies at a sufficiently low temperature, which is called 'zero-point vacancies' (ZPV) [1]. In such a 'supersolid state', non-classical rotational properties are expected, which was argued with emphasis on the connectivity of the wave function [2]. A number of experiments searching for anomalies in thermodynamical or mechanical properties have been performed, but ended with null results [3]. Recently, an abrupt drop in the moment of inertia was found in the torsional oscillation measurements on solid helium 4 confined in a porous media [4] and on a bulk solid helium 4 [5]. This discovery raises the question why other measurements until now have found null results, and leads us to reconsider the definition of superfluidity and that of solids.

A natural way to discuss superfluidity in a confined system is to focus on its rotational properties. Anomalous behaviors of the system are normally attributed to the Bose condensate; in other words, off-diagonal long-range order (ODLRO). The question whether the Bose condensate is a necessary condition for superfluidity has been discussed in terms of academic interest. For solid helium 4, however, this question is not an academic but a practical one. ZPV is still a hypothetical object, but is highly probable if solid helium 4 shows superfluidity [3]. A fundamental feature of crystals lies in their periodicity in density; that is, diagonal long-range order (DLRO). One has to face a serious question whether crystals remain stable while showing superfluidity that violates their periodicity.

In this paper, we assume the existence of ZPV obeying Bose statistics, but do not assume its Bose condensate [6]. Since vacancies are defects on the crystal lattice, it is unlikely that crystals with a macroscopic number of defects remain stable [7]. Rather, in light of the definition of solids, a more natural idea is that the number of ZPV is large, but not macroscopically large in a solid helium 4. Hence, a direction to be explored is the possibility that under the influence of Bose statistics a microscopic number of ZPV's create the non-classical rotational inertia without ODLRO. For the dynamical properties such as superfluidity, *the repulsive interaction generally enhances singular properties due to Bose statistics.* When the container slowly rotates at low temperature, some microscopic regions emerge in solids, in which ZPV and surrounding atoms decouple from the motion of container, thus remaining at rest. The repulsive ZPV's are likely to spread uniformly in coordinate space. This feature makes ZPV outside of these regions behave similarly with ZPV at rest. Otherwise, the density of ZPV would become locally high, thus raising the interaction energy. *The mechanical behavior of microscopic fractions of solids due to Bose statistics (decoupling) is developed to a macroscopic phenomenon by the repulsive interaction.* In this paper, we stress that the non-classical rotational inertia (NCRI) is possible even in the case of no condensate by such a mechanism, and propose its formalism [8].

Zero-point vacancies. Vacancies are usually regarded as localized objects which occasionally move from one position to another in crystals. It takes an energy ϵ_1 for an atom to diffuse to the surface. Further, the lattice distortion due to the vacancy raises the energy by ϵ_2. In quantum crystals, however, the delocalization of the vacancy due to the quantum-mechanical fluctuation lowers the energy by Δ. Hence, it takes $\epsilon_0 = \epsilon_1 + \epsilon_2 - \Delta$ to make a vacancy in the crystal.

In quantum crystals, there is a case in which Δ slightly exceeds $\epsilon_1+\epsilon_2$, thus leading to $\epsilon_0 < 0$. The possible states of such a vacancy are classified by the quasi momentum p. With the tight-binding model applied to ZPV, its energy spectrum has a form as $\epsilon(p) = \epsilon_0 + p^2/2m^*$, where m^* is an effective mass of the vacancy involving surrounding atoms in its motion.

When one vacancy approaches another in crystals, a lattice-distortion pattern by one vacancy is normally not consistent with that by another vacancy. (An atom can not follow two displacement patterns that contradict each

*e-mail address: koh@cc.kochi-u.ac.jp

other.) This situation results in an effective repulsive interaction U between the two vacancies.

Let us consider a crystal with ZPV as $H = H_0 + H_Z$ where H_0 represents a hamiltonian of a crystal with no vacancies, H_Z a hamiltonian of ZPV

$$H_Z = \sum_p \epsilon(p)\Phi_p^\dagger \Phi_p + U \sum_{p,p'} \sum_q \Phi_{p-q}^\dagger \Phi_{p'+q}^\dagger \Phi_{p'} \Phi_p, \quad (1)$$

and Φ denotes an annihilation operator of ZPV as a spinless boson [9]. The motion of vacancies is expressed by Green's function as $G(\omega, p) = 1/(\omega - \epsilon(p))$. Multiple scattering between vacancies is expressed by t-matrix as $t = U/(1 + UG)$. In the tight-binding picture of ZPV, the band width of ZPV has an order of Δ, thus $G \simeq \Delta^{-1}$. As $U \to \infty$, t approaches $1/G \simeq \Delta$. Hence, an energy $\hat{\epsilon}$ of the repulsive ZPV takes a form such as $\hat{\epsilon} = -|\epsilon_0| + n\Delta$, where n is a number density of ZPV. The increase in the number of ZPV stops when $\hat{\epsilon}$ reaches zero, that is, $n = |\epsilon_0|/\Delta$, following which the number of ZPV may change only through their fusion to the surface of crystal. Its relaxation time is much longer than the time required to establish equilibrium for a given number of ZPV. At low temperature, ZPV behaves as a repulsive Bose quasiparticle in the equilibrium state.

For the stability of solids, even in the case of ZPV ($\epsilon_0 < 0$) Δ only slightly exceeds $\epsilon_1 + \epsilon_2$, which would otherwise lead to the collapse of a solid. Because of $|\epsilon_0| = |(\epsilon_1 + \epsilon_2) - \Delta| \ll \Delta$, the number density satisfies $n = |\epsilon_0|/\Delta \ll 1$. Hence, it is unlikely that the number of ZPV is a macroscopic one.

Moment of inertia. By the torsional oscillator experiment, one measures the moment of inertia I of a sample as a response to the restoring force of the torsion rod. As long as the external force is small, I is determined by the rotational property of the system without that force.

Consider a solid helium 4 in a uniform rotation round z-axis with angular velocity Ω. A hamiltonian in a coordinate system rotating with the body is $H - \boldsymbol{\Omega} \cdot \boldsymbol{L}$, where $\boldsymbol{L}$ is the total angular momentum. The rotation is equivalent to the application of the probe acting on the sample. The perturbation $-\boldsymbol{\Omega} \cdot \boldsymbol{L}$ is cast in the form $-\sum_i (\boldsymbol{\Omega} \times \mathrm{r}) \cdot \boldsymbol{p}$, where $\boldsymbol{\Omega} \times \boldsymbol{r} \equiv \boldsymbol{v}_d(\boldsymbol{r})$ is the drift velocity of the sample at point $\boldsymbol{r}$. With the current density $\boldsymbol{J}(\boldsymbol{r})$, the perturbation has a form as

$$-\boldsymbol{\Omega} \cdot \boldsymbol{L} = -\int \boldsymbol{v}_d(\boldsymbol{r}) \cdot \boldsymbol{J}(\boldsymbol{r}) d\boldsymbol{r}. \quad (2)$$

Because of $div \boldsymbol{v}_d(\boldsymbol{r}) = 0$, Eq.(2) says that the rotation represented by $\boldsymbol{v}_d(\boldsymbol{r})$ acts as a transverse-vector probe to the excitations in solids [10]. We need the linear response of the system $\boldsymbol{J}(\boldsymbol{r})$ to $\boldsymbol{v}_d(\boldsymbol{r})$. Since a solid normally rotates like a rigid body, we can use the mass density ρ in $\boldsymbol{J}(\boldsymbol{r}) = \rho \boldsymbol{v}_d(\boldsymbol{r})$. Microscopically, however, one must begin with the transverse susceptibility $\chi^T(q, \omega)$ of the system. (By definition, the mass density ρ is regarded as the longitudinal response to a force as $\rho = nm = \chi^L(0, 0)$.) One assumes the spatial homogeneity of the sample during rotation. Hence, $\mathbf{J}(\mathbf{r})$ is approximated as

$$\boldsymbol{J}(\boldsymbol{r}) = \left[\lim_{q \to 0} \chi^T(q, 0)\right] \boldsymbol{v}_d(\boldsymbol{r}) \quad (3)$$

Using Eq,(3) and $\boldsymbol{v}_d = (-\Omega y, \Omega x, 0)$ in the right-hand side of Eq.(2), and $\boldsymbol{\Omega} = (0, 0, \Omega)$ in its left-hand side, one obtains the angular momentum L_z, hence the moment of inertia $I_z = L_z/\Omega$ as

$$I_z = \chi^T(0, 0) \int_V (x^2 + y^2) d\mathbf{r}. \quad (4)$$

(A) The susceptibility of a normal solid satisfies $\chi^T(0, 0) = \chi^L(0, 0)$. Hence, replacing $\chi^T(0, 0)$ by ρ in Eq.(4), one obtains the definition of classical I_z^c.

The generalized susceptibility is defined as

$$\chi_{\mu\nu}(q, \omega) = \frac{q_\mu q_\nu}{q^2} \chi^L(q, \omega) + \left(\delta_{\mu\nu} - \frac{q_\mu q_\nu}{q^2}\right) \chi^T(q, \omega). \quad (5)$$

For the later use, we define a term proportional to $q_\mu q_\nu$ in $\chi_{\mu\nu}$ as $\hat{\chi}_{\mu\nu}$ so that

$$\begin{aligned} \chi_{\mu\nu}(q, \omega) &= \delta_{\mu\nu} \chi^T(q, \omega) + q_\mu q_\nu \left(\frac{\chi^L(q, \omega) - \chi^T(q, \omega)}{q^2}\right) \\ &\equiv \delta_{\mu\nu} \chi^T + \hat{\chi}_{\mu\nu}(q, \omega), \end{aligned} \quad (6)$$

and rewrite Eq.(4) as

$$I_z = I_z^c - \lim_{q \to 0} \left[\frac{q^2}{q_\mu q_\nu} \hat{\chi}_{\mu\nu}(q, 0)\right] \int_V (x^2 + y^2) d\mathbf{r}. \quad (7)$$

(B) At the onset of ZPV, according to $H = H_0 + H_Z$, the susceptibility $\chi(q, \omega)$ splits into $\chi_0(q, \omega)$ and $\chi^{ZPV}(q, \omega)$ [11]. The normal part gives no contribution to NCRI in Eq.(7) due to $\chi_0^L(0, 0) = \chi_0^T(0, 0)$, whereas the validity of $\chi^{ZPV,L}(0, 0) = \chi^{ZPV,T}(0, 0)$ must be examined. For the appearance of NCRI, the balance between the longitudinal and the transverse excitation must be destroyed on a macroscopic scale ($q \to 0$). On this point, one can trace back to Feynman's physical argument on how the Bose-statistical coherence suppresses the transverse excitation, and destroys this balance [12]. In his argument, not the Bose condensate but Bose statistics is essential.

(C) To see the effect of the repulsive interaction, let us consider $\chi_{\mu\nu}$ of ZPV as an ideal Bose gas. Within the linear response, it is defined as

$$\begin{aligned} &\chi_{\mu\nu}^{ZPV}(q, \omega_n) \\ &= \frac{1}{V} \int_0^\beta d\tau \exp(i\omega_n \tau) < S|T_\tau J_\mu(q, \tau) J_\nu(q, 0)|S > , \end{aligned} \quad (8)$$

where $J_\mu(q, \tau)$ is a current of ZPV ($\hbar = 1$) [13]

$$J_\mu(q,\tau) = \sum_{p,n} \left(p + \frac{q}{2}\right)_\mu \Phi_p^\dagger \Phi_{p+q} e^{i\omega_n \tau}, \tag{9}$$

and $|S>$ is a ground sate of $H_0 + \sum_p \epsilon(p)\Phi_p^\dagger\Phi_p$. Its $\hat{\chi}_{\mu\nu}(q,\omega)$ is given by

$$\hat{\chi}_{\mu\nu}^{ZPV}(q,\omega) = -\frac{q_\mu q_\nu}{4} \sum_p \frac{f(\epsilon(p)) - f(\epsilon(p+q))}{\omega + \epsilon(p) - \epsilon(p+q)}, \tag{10}$$

where $f(\epsilon(p))$ is the Bose-Einstein distribution.

(1) If ZPV would form the condensate, $f(\epsilon(p))$ in Eq.(10) is a macroscopic number for $p = 0$ and nearly zero for $p \neq 0$. Thus, in the sum over p in the right-hand side of Eq.(10), only two terms corresponding to $p = 0$ and $p = -q$ remain, with a result that

$$\hat{\chi}_{\mu\nu}^{ZPV}(q,0) = m^* n_0 \frac{q_\mu q_\nu}{q^2}, \tag{11}$$

where n_0 is the number density of the condensate. Equation.(7) with Eq.(11) shows NCRI. (The relative degree $(I_z - I_z^c)/I_z^c$ of the deviation from the classical moment of inertia would be simply proportional to the number density of the condensate.)

(2) When ZPV forms no condensate, however, the sum over p in Eq.(10) is carried out by replacing it with an integral, and one notices that q^{-2} dependence disappears in the result. Hence, without the repulsive interaction, BEC is a necessary condition of NCRI.

<u>*Non-classical rotational inertia.*</u> We will formulate a mechanism of NCMI under the more realistic condition for a solid helium 4. Instead of $|S>$ in Eq.(8), a ground state $|G>$ of $H_0 + H_Z$ must be used as

$$< G|T_\tau J_\mu(x,\tau) J_\nu(0,0)|G > = \frac{< S|T_\tau \hat{J}_\mu(x,\tau)\hat{J}_\nu(0,0) exp\left[-i\int_0^{1/\beta} d\tau H_I(\tau)\right]|S >}{< S|exp\left[-i\int_0^{1/\beta} d\tau H_I(\tau)\right]|S >}, \tag{12}$$

where $H_I(\tau)$ represents the repulsive interaction. Due to the repulsive interaction U contained in $exp(-i\int H_I(\tau)d\tau)$, the scattering between ZPV's frequently occurs as illustrated by an upper bubble with a dotted line in Fig.1(a). The current-current response tensor $J_\mu(x,\tau)J_\nu(0,0)$ is depicted by a lower bubble.

Since $|G>$ is a ground state under the influence of Bose statistics, the perturbation must be developed in such a way that as the order of the expansion increases, the susceptibility gradually includes a new effect due to Bose statistics. An important feature is that ZPV in $J_\mu(x,\tau)J_\nu(0,0)$ and ZPV in the upper bubble due to the repulsive interaction form a coherent wave function as a whole. Hence, we must seriously consider the influence of Bose statistics on the graph like Fig.1(a). When one of the two ZPV's in the lower bubble and that in the upper bubble have the same momentum $(p = p')$, and the other ZPV in the two bubbles have the same momentum $(p + q = p' + q')$, a graph made by interchanging these two types of ZPV's must be included in the expansion of $\chi_{\mu\nu}^{ZPV}$. The interchange of ZPV lines with p and $p'(= p)$ in Fig.1(a) yields Fig.1(b). Further, the interchange of ZPV lines with $p + q$ and $p + q'(= p + q)$ yields Fig.1(c), a graph linked by the repulsive interaction, whose contribution to $\chi_{\mu\nu}^{ZPV}$ is given by

$$\chi_{\mu\nu}^{ZPV,(1)}(q,\omega) = U \sum_p (p + \frac{q}{2})_\mu (p + \frac{q}{2})_\nu \left[-\frac{f(\epsilon(p)) - f(\epsilon(p+q))}{\omega + \epsilon(p) - \epsilon(p+q)}\right]^2. \tag{13}$$

With decreasing temperature, the coherent wave function grows to a large size, and the interchange of ZPV due to Bose statistics like Fig.1 occurs many times. Hence, one can not ignore the higher-order term $\chi_{\mu\nu}^{ZPV,(n)}$ which is significant in the larger coherent wave function. Among many ZPV's with various momentums, ZPV's at rest with $p = 0$ play a key role, leading to the following form

$$\hat{\chi}_{\mu\nu}^{ZPV,(n)}(q,0) = \frac{q_\mu q_\nu}{4} U^n F_\beta(q)^{n+1}, \tag{14}$$

where

$$F_\beta(q) = \frac{(\exp(-\beta\mu) - 1)^{-1} - (\exp(\beta[\epsilon(q) - \mu]) - 1)^{-1}}{\epsilon(q)}, \tag{15}$$

and μ is a chemical potential. $F_\beta(q)$ is a positive monotonously decreasing function of q^2 which approaches zero as $q^2 \to \infty$. As $\mu \to 0$, $F_\beta(q)$ increases at any q. An expansion form of $F_\beta(q)$ around $q^2 = 0$ is given by

$$F_\beta(q) = \frac{\beta \exp(\beta\mu)}{(1 - \exp(\beta\mu))^2} - \frac{\beta^2}{2} \exp(\beta\mu) \frac{(1 + \exp(\beta\mu))}{(1 - \exp(\beta\mu))^3} \epsilon(q) + \cdots. \tag{16}$$

When $q^2 \hat{\chi}_{\mu\nu}/q_\mu q_\nu$ has a finite value at $q \to 0$ in Eq.(7), that is, when a power series in U

$$\hat{\chi}_{\mu\nu}^{ZPV}(q,0) = \frac{q_\mu q_\nu}{4} \sum_{n=0}^{\infty} U^n F_\beta(q)^{n+1}, \tag{17}$$

diverges as q^{-2} at $q \to 0$, the moment of inertia shows a non-classical behavior. In normal solids ($\beta\mu \ll 0$), a small $F_\beta(q)$ guarantees the convergence of $\hat{\chi}_{\mu\nu}^{ZPV}(q,0)$, with a result that,

$$\hat{\chi}_{\mu\nu}^{ZPV}(q,0) = \frac{q_\mu q_\nu}{4} \frac{F_\beta(q)}{1 - UF_\beta(q)}. \tag{18}$$

With decreasing temperature ($\mu \to 0$), however, a gradual increase of $F_\beta(q)$ makes the higher-order term significant in Eq.(17), finally leading to the divergence of power

series in $\hat{\chi}^{ZPV}_{\mu\nu}(q,0)$. The convergence condition is first violated at $q=0$ when $UF_\beta(0)=1$, that is,

$$U\beta = 4\sinh^2\left(\frac{\beta\mu}{2}\right). \tag{19}$$

From now, we call T_0 satisfying Eq.(19) an onset temperature of NCRI. Equation (19) implies that *a solid with ZPV shows NCRI without the condensate* ($\mu \neq 0$). At $T=T_0$, substituting Eq.(16) in Eq.(18) with the aid of Eq.(19), we find as a leading term

$$\hat{\chi}^{ZPV}_{\mu\nu}(q,0) = \frac{m^*}{2\sinh|\beta_0\mu|}\frac{q_\mu q_\nu}{q^2}. \tag{20}$$

In view of Eq.(6), Eq.(20) leads to $\chi^{ZPV,L}(q,0) \neq \chi^{ZPV,T}(q,0)$ at $q \to 0$. Since the large-scale interchange of bosons plays an essential role for deriving Eq.(20), this result confirms Feynman's intuitive argument on the influence of Bose statistics on the excitations. Using Eq.(20) in Eq.(7) with the aid of Eq.(19), we obtain

$$I_z = I_z^c - \frac{m^*}{\sqrt{\frac{U}{k_BT_0}\left(4+\frac{U}{k_BT_0}\right)}}\int_V (x^2+y^2)d\mathbf{r}, \tag{21}$$

which implies that $(I_z - I_z^c)/I_z^c$ for superfluidity by ZPV does not allow the one-body interpretation as ρ_s/ρ, but reflects the many-body effect due to Bose statistics and the repulsive interaction. Since ZPV is not a particle in the literal sense, but represents the motion of surrounding atoms, this is a natural result.

Discussion. Superfluidity is a complex of phenomena, and therefore has some different definitions such as, (1) persistent current without friction, (2) the Hess-Fairbank effect, (3) quantized circulation, (4) almost no friction on moving objects in the system below the critical velocity, (5) peculiar collective excitations and (6) the Josephson effect [14]. (Strictly speaking, the experiment by Kim and Chan adds a new definition of superfluidity to the list.) Since these phenomena (1) $\sim$ (6) are observed to occur together in most materials exhibiting superfluidity (including superconductors), it is difficult to elucidate the exact nature of the relationships between these definitions. From the standpoint of this paper, there is no reason to expect that all superfluid-like behaviors corresponding to these definitions should occur in quantum crystals. We must therefore classify them into two categories. The first is the phenomenon which an interplay of Bose statistics and the repulsive force creates without the condensate, and the second is that for which the condensate is necessary. The first and the second corresponds to an observable and an unobservable phenomenon in quantum crystals, respectively. (From this viewpoint, we must put a new interpretation on some 'null results' in earlier measurements of mechanical properties of a solid helium 4 [15].) We can regard a solid helium 4, together with a helium 4 film [16], as appropriate material for examining the relationship between the different definitions of superfluidity.

[1] A.F.Andreef and I.M.Lifshitz, Zh.Eksp.teor.Fiz. **56**, 2057(1969), [Sov.Phys.JETP. **29**, 1107(1969)].

[2] A.J.Leggett, Phys.Rev. Lett.**25**,1543(1970).

[3] As a review, M.W.Meisel, Physica. B**178**, 121(1992).

[4] E.Kim and M.H.W.Chan, Nature. **427**, 225(2004).

[5] E.Kim and M.H.W.Chan, Science. **305**, 1941(2004).

[6] Although the Bose condensate is a most peculiar feature of Bose statistics, more fundamental is a fact that particles obey Bose statistics even when BEC is absent.

[7] This view agrees with the strict upper bounds to the density of ZPV derived from the null results.

[8] The Meissner effect is the counterpart of NCRI in the charged Bose systems. Its appearance prior to BEC was formulated in S.Koh, Phy.Rev,B, **68**, 144502(2003).

[9] If the defects are dislocations, the model must be improved to deal with the extended objects.

[10] As a review, P.Nozieres, in *Quantum Fluids* (ed by D.E.Brewer), 1 (North Holland, Amsterdam, 1966).

[11] It is assumed that the motion of vacancies can be separately considered from other excitations in crystals.

[12] R.P.Feynman, in *Progress in Low Temp Phys***1**, (ed C.J.Gorter), 17 (North-Holland, Amsterdam, 1955).

[13] Since $\boldsymbol{v}_d$ is the neutral counterpart of a vector potential A_μ, Eq.(9) has the same structure as that in the charged Bose gas.

[14] A.J.Leggett, in *Bose-Einstein condensation*, (ed by C.M.Savage and M.P.Das), 1 (World Scientific, 2000).

[15] Since the condensate is absent, there is little probability of detecting signs in thermodynamical properties such as the temperature dependence of pressure.

[16] D.J.Bishop and J.D.Reppy, Phys.Rev.Lett. **40**, 1727 (1978).

FIG. 1. The first-order Feynman diagram of the current-current response tensor. The black and the white small circle represents a vector and a scalar vertex, respectively. The dotted line denotes the repulsive interaction.